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U.S. DEPARTMENT OF THE INTERIOR
SAFETY EFFICIENCY
MINERAL INDUSTRIES
BUREAU OF MINES

STUDIES ON DEFLAGRATION TO DETONATION IN PROPELLANTS AND EXPLOSIVES

QUARTERLY REPORT

January 1, 1961 to March 31, 1961

BUREAU OF MINES, PITTSBURGH, PA.

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Prepared for:

Advanced Research Projects Agency
Office of the Secretary of Defense
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STUDIES ON DEFLAGRATION TO DETONATION
IN PROPELLANTS AND EXPLOSIVES

SUMMARY

During the quarter, the U. S. Naval Propellant Plant high-performance double-base propellant was tested under extreme confinement. Thus far it has been impossible to attain a transition from deflagration to detonation in this material, in spite of the fact that the confined critical diameter is only about 1.5 cm. when directly initiated by explosive-generated shock waves.

Some basic experiments have been conducted, using simple probe systems to determine electron and ion activity in the reaction zone of a detonating explosive. Ultimately it may be possible to prevent a transition by applying externally generated electric and/or magnetic fields. This is important because the trend is toward the use of more energetic materials for which control of the physical parameters may, in the future, not suffice.

A pressure probe system for the continuous display of compression wave velocities was developed and applied to delayed detonation in explosives as well as to pressure waves in an inert liquid body. The indications are that such a system may be used effectively to reveal pre-detonation pressure waves in opaque solids under heavy confinement.

INTRODUCTION

One objective of this investigation is to study the mechanism by which propellants and explosives go from deflagration to detonation. Another is to determine the detonation sensitivity of current and developmental propellants when subjected to shock waves and relatively mild thermal stimuli. Current solid propellants are like most solid explosives in that they can be detonated by direct shock-wave initiation by another explosive. However, the manner in which a deflagration develops into a detonation is not completely understood, although hypotheses have been advanced and mechanisms proposed. A recent paper by N. Griffiths and J. M. Groocock[1/] suggests some details of the deflagration-to-detonation event. Their work deals with narrow columns of highly confined pure explosives. It is postulated that during their combustion a thermal explosion occurs behind an accelerating chemical reaction causing the transition to detonation. This is similar, but not completely analogous, to the mechanism for initiation proposed by K. K. Andreev[2/] in which the explosion behind the propagation front is attributed to "explosion-like" burning of explosive particles suspended in the burning products. The hypothesis of Griffiths and Groocock for solids is supported by gaseous detonation studies conducted at the Bureau of Mines by Greifer, et al.[3/] about 1956.

1/ Griffiths, N. and Groocock, J. M., "The Burning to Detonation of Solid Explosives", preprinted from the Jour. of the Chemical Society, November 1960 (814), pp. 4154-4162.

2/ Andreev, K. K., "Some Considerations on the Mechanism of Initiation of Detonation in Explosives", Proceedings of the Royal Society, No. 1245, Vol. 246, July 29, 1958.

3/ Greifer, B., Cooper, J. C., Gibson, F. C., and Mason, C. M., "Combustion and Detonation in Gases", reprinted from Jour. of Applied Physics, Vol. 28, No. 3, March 1957, pp. 289-294.

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The latter report that compression waves which travel ahead of the combustion wave steepen into a shock wave. As the process accelerates and the distance between the waves decreases, local explosions occur in the unreacted material ever closer to the shock front. When such an explosion occurs immediately behind the shock front, sufficient energy is transmitted to the shock wave and steady-state detonation ensues. Obviously, such studies are best adapted to transparent systems where non-reactive shocks can be displayed by photographic techniques; however, combining pressure-type probes with ionization-type probes will provide improved techniques for studies on propellants and explosives that are opaque and are to be tested under heavy confinement.

Control of the physical properties that influence detonability may suffice for the present; but since propellants are becoming more and more energetic, means other than control of physical parameters may be required. Ultimately, electrical control for the suppression of detonation may offer a solution to this problem.

EXPERIMENTAL

A. Experimental work on confined propellant grains

The investigation of transition from deflagration to detonation in propellants was continued, employing self-sealing pressure vessels of the type described in the previous report[4/]. Each successive experiment in a series was modified to minimize or eliminate the shortcomings of the previous one. In an early

4/ Bureau of Mines Quarterly Report, "Studies on Deflagration to Detonation in Propellants and Explosives", Office of the Secretary of Defense, ARPA Order No. 44-61, October 1, 1960 to December 31, 1960.

experiment in a cold rolled steel vessel, the gaseous combustion products were contained prior to instantaneous rupture of the vessel which burst into six large fragments. However, the rupture occurred after only a relatively small amount of propellant had burned and many small pieces of unburned propellant were recovered. In this case the structural weakness of the vessel had limited the development of compression waves sufficient to establish a detonation.

Subsequently, vessels were made of high-alloy steel with a minimum tensile yield of 75,000 psi. These vessels were designed to provide a factor of about two in strength over the cold rolled and to remain intact long enough to permit a pressure buildup that would be more likely to cause a detonation in the burning propellant. The propellant ignitor was located at one end and the instrumentation leads passed through sealed ports at both ends of the vessel. A wire strain gage was attached to the outer wall of the vessel to measure the circumferential strain (which qualitatively relates to the internal pressure) and a resistance wire element was placed on the axis of the propellant charge to measure propagation velocity. A vessel of this type remained intact but the gaseous products escaped through the port at the ignitor end. As the streaming products are very erosive, even a minute leakage quickly enlarges the escape hole and appreciably limits the amplitude of the internal compression wave. Although the pressure developed was insufficient to rupture the vessel, the external gage indicated a strain of 2200 μin./inch (200,000 psi. internal pressure) before leakage of the products gases. After leakage occurred, the propellant combustion products escaped into the atmosphere and the material burned to completion at a rate of about 1.46 cm/sec.

Because containment of the gases at the ignitor end seemed impractical, the vessel loading was modified to provide only a single port in the downstream end of the vessel. Using a vessel of this type, a strain of 3700 μin./inch (340,000 psi. internal pressure) and a burning velocity of as high as 125 m/sec. were obtained; but again internal activity was limited by leakage of the gaseous products at the instrumentation port. In order to increase the rate of pressure rise and minimize the possibility of early leakage, it was felt that the propellant ignitor should be positioned on the axis and near the center of the propellant charge. Although this reduced the run-up length, the amplitude of the compression wave would be enhanced because of the greater burning surface area. In an experiment of this type, sufficient pressure had developed when the burning velocity reached 37 cm/sec., as shown by the resistance element record, to cause the propellant material and instrumentation leads to be extruded from the downstream port. This interrupted the velocity measurements but the propellant burned for about 30 milliseconds longer. The peak internal pressure produced was estimated to be about 500,000 psi.

To determine whether this type of vessel could contain a sufficient pressure impulse to cause a transition from deflagration to detonation in a high explosive, a similar vessel was filled with RDX having a density of 1.23 gm/cm^3. The ignitor -- an electrically heated wire imbedded in about a 1/2 gram of the double-base propellant -- was placed in the body of the explosive about one inch from the end of the vessel. Before the vessel leaked, a transition to detonation occurred. An oscilloscope record of the latter portion of the event (Figure 1) shows a high-order detonation with a velocity of 6850 m/sec., which is the hydrodynamic detonation rate for RDX at the density employed.

B. Critical diameter studies on propellant grains

It had been reported earlier[4] that detonation ceased in a 1.25 cm.-diameter segment of a stepped propellant grain that consisted of five cylindrical sections of decreasing diameters; the grain was prepared from a single propellant cylinder and shock initiated with a high explosive. This experiment was repeated, keeping the diameters constant but using an ℓ/d ratio that was considerably larger, to determine if the position of the diameter discontinuity influenced the region where detonation ceased. As in the earlier experiment, the detonation failed after propagating about 4 mm. into the 1.25 cm.-diameter segment of the grain. The grain configuration and record are shown in Figure 2; the axial resistance element has a section of its length shorted out to provide a fiducial mark on the oscilloscope trace from which detonation zone position vs. time can be accurately measured. The point of cessation as determined from the oscillogram agreed well with the length of the propellant strand recovered after the test. Thus it appears that a critical diameter for detonation in this high-performance double-base propellant is less than 1.7 cm. and more than 1.25 cm. or perhaps about 1.5 cm.

C. Experimental work on electrical effects that accompany detonation

A review of the literature indicates that detonation in certain gaseous mixtures was arrested by the application of an axial electric field. This experiment, along with others using magnetic fields, was described in a paper by Bone, Frazer and Wheeler[5/] in 1936, although the work had been proposed in 1909 by J. J. Thompson in a discussion on gaseous combustion which, in turn, was based on work of Turpin[6/] reported in 1893. Thompson suggested that the electron activity associated with detonation might precede the detonation by ionization of the unreacted gas ahead of the detonation wave; if this were so, their motions might be influenced by a transverse magnetic field. Bone, et al. concluded that spinning detonation in gaseous mixtures of carbon monoxide and oxygen could be accelerated by strong axial fields and that detonation could be suppressed by axial electrical fields, having a negative to positive potential gradient of 5000 v/cm., into which the detonation passed. Recent research by the Bureau of Mines[7/] on the effects of electric and magnetic fields on detonations in hydrogen-oxygen mixtures show a negligible effect on the propagation velocity as the detonation moves through either external magnetic or electric fields. However, to date the fields have been applied to stable hydrodynamic detonations where the susceptibility to wave front degradation is probably least. It thus appears that prevention of a transition from deflagration to detonation in explosives and propellants may be practical if the appropriate

5/ Bone, W. A., Fraser, R. P., and Wheeler, W. H., Phil. Trans. Roy. Soc. (London) A235, 29 (1936).

6/ Studies from the Physical and Chemical Laboratories of the Owens College, Manchester, Vol. I, pp. 283-95 (1893).

7/ This research on "Electrical and Magnetic Effects on Detonation" is currently being conducted for the Department of the Army, Aberdeen Proving Ground, Maryland.

external field is applied during the pre-detonation-growth stage, providing a charge separation exists in the reaction zone. A few preliminary experiments were conducted to determine if precursor electron activity existed ahead of a detonation front, and to measure the polarity of generated emf's as sensed by a simple probe system. A charge configuration that was used and the resulting oscillogram are shown in Figure 3(a) and (b). No precursor activity is indicated and the time displacement of the waveforms produced on a dual-beam oscilloscope indicate a velocity of detonation anticipated for granular tetryl of pouring density. Essentially symmetrical waveforms are produced at the two-probe stations except that the greater column length provides a longer voltage pulse resulting from a longer contact period with the hot gaseous products cloud. A positive and negative potential scanning time of about a half microsecond is indicated for each. If the measurement is considered the result of a surface effect, a unipotential zone thickness of about 2.5 mm. is indicated. As this appears to be an unreasonable reaction zone thickness, the effective potential gradient must exist between the surface and the axis or be the combined effects of both.

Since the initiation of detonation by non-reactive shock waves and thermal stimuli results from compression wave movement through the reactive medium cannot be mapped by ionization probe techniques, a pressure probe transducer has been developed for continuously determining compression wave velocities. This method is based on a technique of Amster et al.[8/] in which a thin-walled copper tube with an insulated concentric inner lead collapses due to the differential pressure, thus acting as a switch. The

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8/ Amster, Adolph B. and Beauregard, Raymond L., "Pressure Sensing Probes for Detecting Shock Waves", Rev. Sci. Instruments, October 1959, p. 942.

resulting pulses are used to measure incremental velocities by oscillographic or counter-chronograph techniques. This probe is similar to, but much smaller than, a duPont T-1 target -- an electric blasting cap assembly without a bridgewire or explosive -- that is widely used for velocity determination in dynamites where the switch action depends on the collapse of the detonator shell on the bridgewire support leads. The modification of the pressure probe technique for continuous measurement combines the collapsing tube and the resistance element methods. Here a thin-walled aluminum tube encloses a co-axial resistance element and the progressive collapse of the tube by a propagating compression wave causes the resistance element to be electrically shorted with a resultant decrease in voltage drop across the intact portion of the resistance element. Display of this voltage with respect to time permits the determination of position and velocity of the compression wave if the front of the wave is normal to the tubing.

Exploratory trials were made on the performance of probe elements of this type in both water and granular explosive systems. An oscillogram of the compression wave velocity in water is shown in Figure 4; the distance on the ordinate is 2.1 cm/div. and the time on the abscissa is 5 μsec/div. The record shows the transition from the overdriven shock wave into a sonic wave 3.8 cm. from the barrier; a steady-state velocity of 1.4 mm/μsec. was maintained and measured until the pressure was no longer sufficient to collapse the tubing.

This technique has been applied to tests with an explosive to compare the compression wave position and the velocity with that of the ionized region in the detonation front, as determined by the resistance element method. A pressure probe was placed in a tetryl charge parallel to the axis, at a distance of 8 mm. from a resistance element, as shown in Figure 5(a). The results were recorded simultaneously on a dual-beam oscilloscope having an equal sweep speed for both traces and on separate oscilloscopes which give improved accuracy in trace analysis. The dual-beam sweep was 5 μsec./div. (Figure 5(b); identical slopes are not expected because the lineal resistance for the pressure probe element (upper trace) is 21.6 ohms/cm. and the ionization probe (lower trace) is 11.73 ohms/cm. The velocity through the rubber barrier between the donor and an acceptor is about 3 mm/μsec. and identical results in both measuring systems were obtained. The ionization probe shows a period of 12 μsec. during which ionization did not exist or was insufficient to cause conduction in the wire element probe. However, during this period the pressure probe showed a steady compression wave velocity of 1.7 mm/μsec. that followed a 3 μsec. interval during which a velocity of about 1 mm/μsec. was recorded; this relatively low value may be due to interface effects. At a point where ionization first appeared an acceleration of the compression wave developed, becoming a steady-state propagation when the ionization probe indicated that the hydrodynamic velocity had been achieved; the terminal velocity was about 5.2 mm/μsec. for each method. The oscillogram of the pressure probe record on an expanded time base is shown in Figure 5(c), and the ionization probe record in Figure 5(d). The time base on the abscissa is 5 μsec./div. in both cases and the displacement on the ordinate is 3.2 cm/div. and 4.0 cm/div., respectively. It thus appears that the pressure probe system can be applied in studies of weak thermal initiation of confined propellant grains to plot the rate of movement and development of compression waves prior to transition to detonation. A better understanding of the probe systems can be achieved when high-speed photographic equipment, now on order, becomes available.

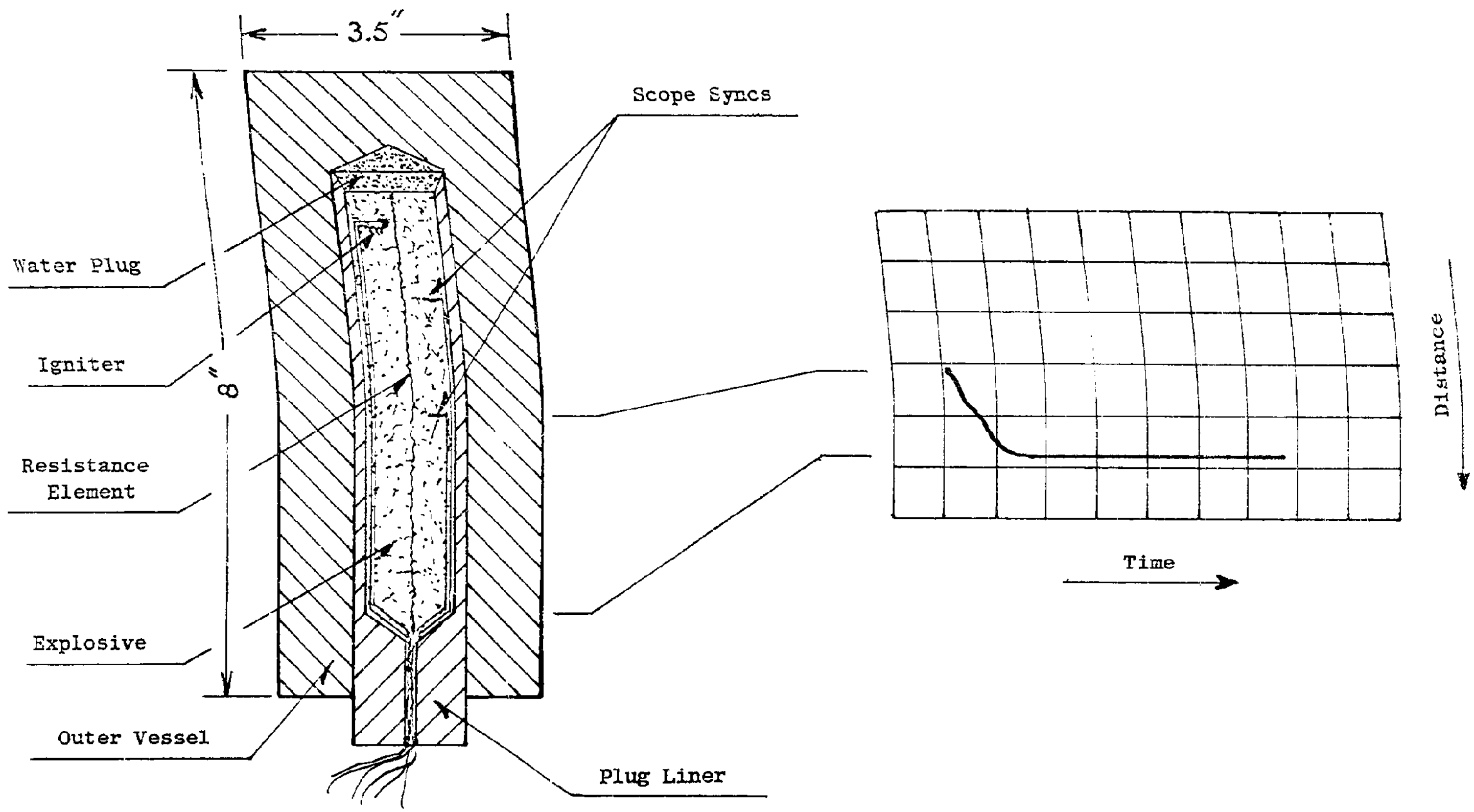


Figure 1. - Test configuration used to produce a deflagration-to-detonation transition in RDX (density 1.23 g/cm^3). The oscillogram indicates a terminal velocity of 6.9 mm/μsec. but the point of transition is not shown because the oscilloscope sync was located in a region where steady-state detonation had already existed. Vessel configuration is identical to that used for detonability studies on high-performance double-base propellant grains where transitions have not yet been obtained.

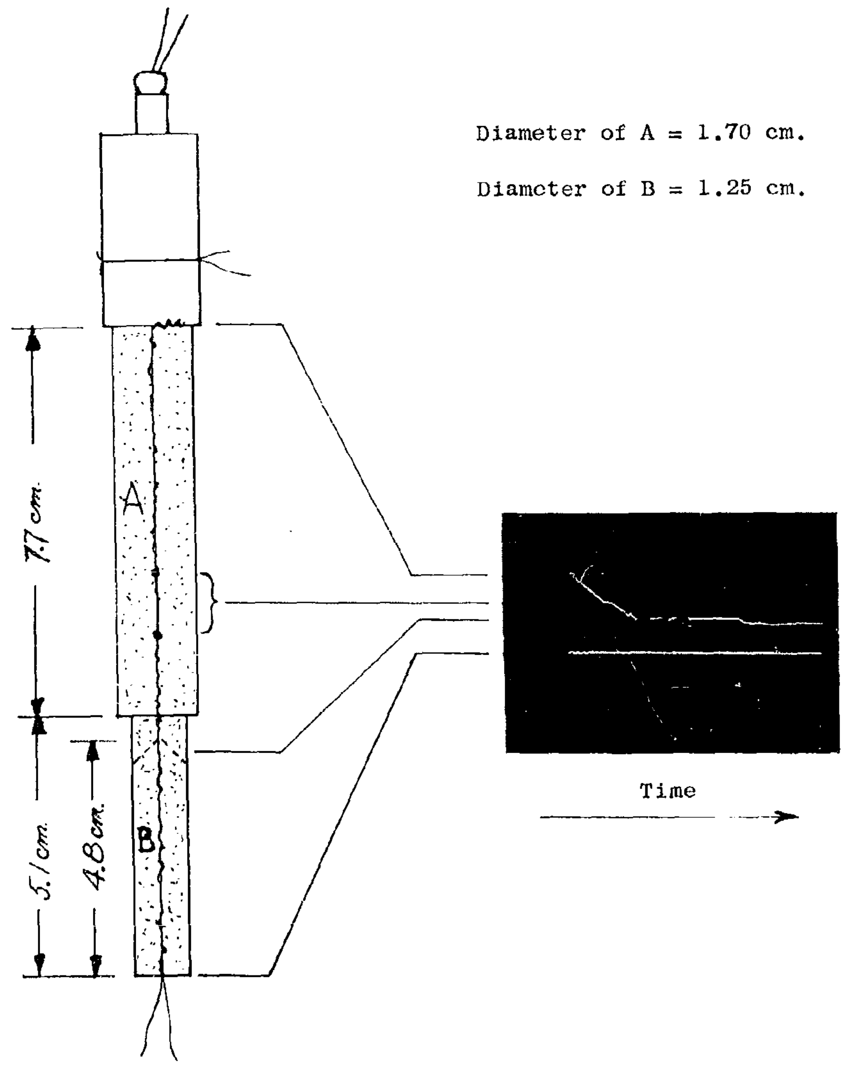


Figure 2. - Charge configuration and oscillogram resulting from a detonation in a critical diameter-type propellant grain. Detonation ceased in the 1.25 cm.-diameter segment. The oscillogram shows a steady-state velocity of about 6.5 mm/μsec. that decays to about 5 mm/μsec. as the detonation approaches the diameter discontinuity. The horizontal section about midway on the trace is the effect of shorted turns in the resistance element and is used as a fiducial marker.

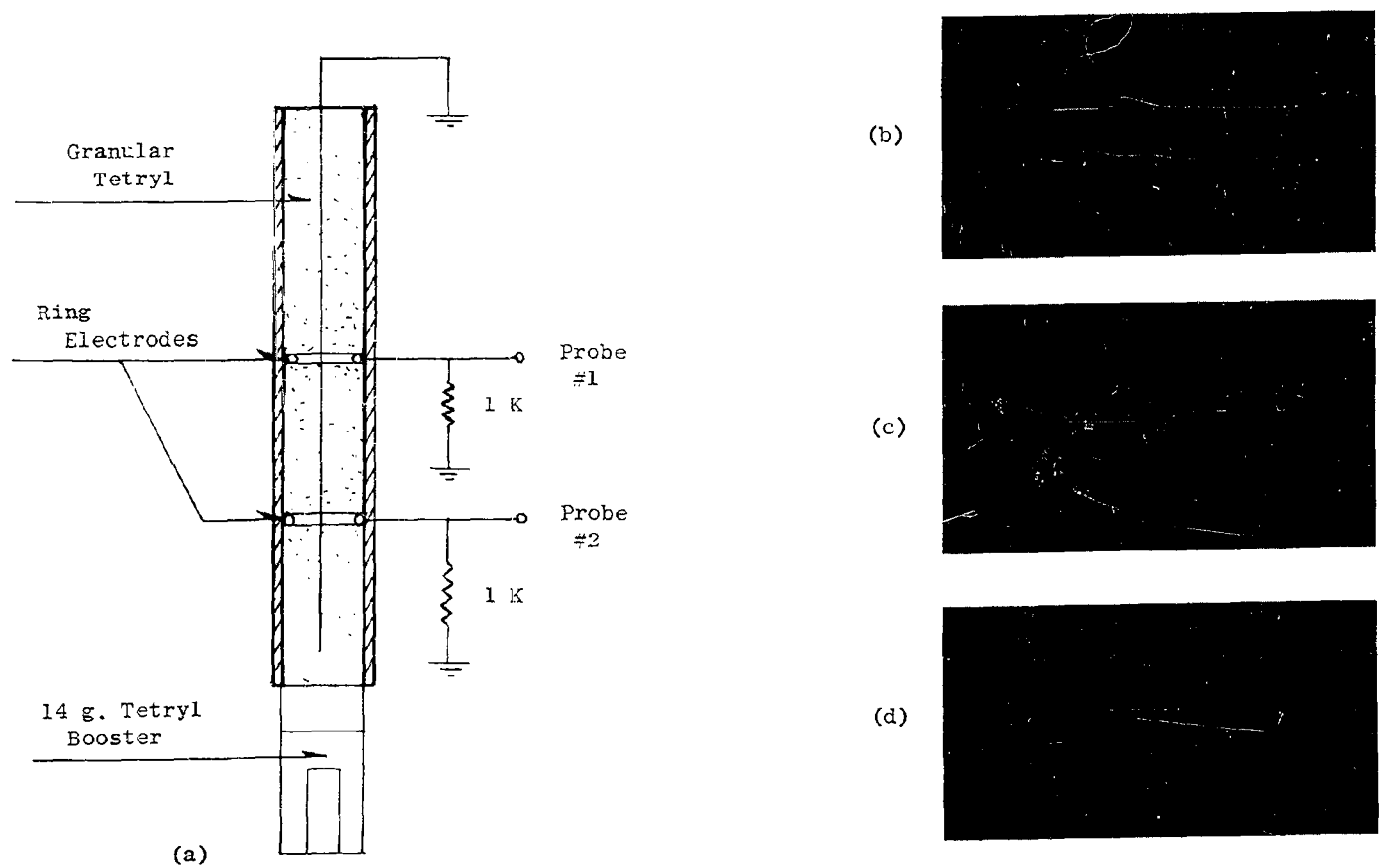


Figure 3. - Charge configuration (a) showing the probe system and oscillograms (b), (c) and (d) obtained showing electrical activity in detonating tetryl. The emf's generated are shown in the dual-beam oscillogram (b) where the lower trace is the upstream of the first probe. The time base is 5 μsec./div. and increases from left to right. An expanded time base showing emf's generated for the first and second probes are shown in (c) and (d) respectively, where respective sweep times are 1 μsec./div. and 2 μsec./div. Based on waveforms (rate of rise of the initial pulse) obtained, no precursor activity is indicated.

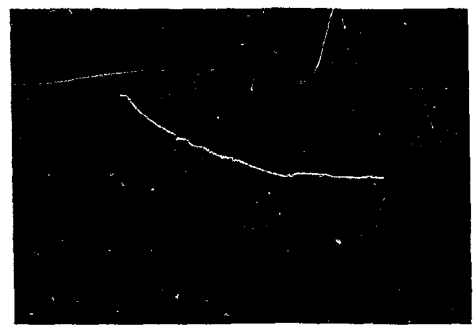
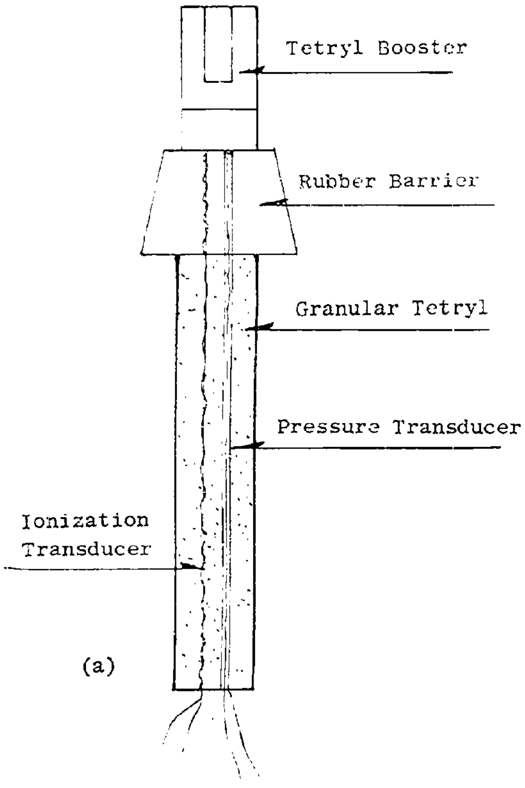
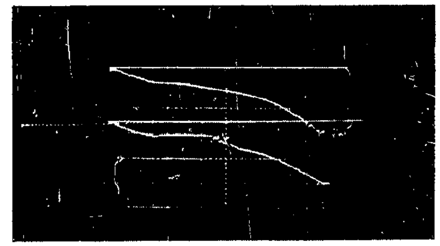


Figure 4. - Oscillogram showing an explosive-produced compression wave passing through a column of water. A pressure-sensitive transducer consisting of a resistance element of the type used for continuous determination of velocities of detonation is placed co-axially in a thin-walled aluminum tube (wall thickness 1.5 mils and o.d. 26 mils). Collapse of the tubing progressively shortens the electrical length of the resistance element and the voltage across the "intact" portion of the element with respect to time represents the position of the wave. The terminal or sonic velocity of the wave is 1.4 mm/μsec. Discontinuities on the trace when the overdriven wave becomes sonic are probably due to compression wave interactions on the axis of the cylindrical vessel.



(a)



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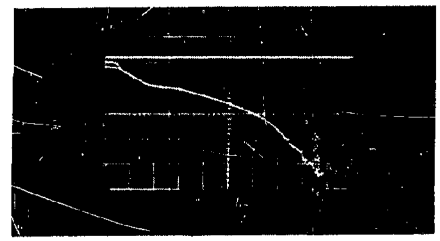
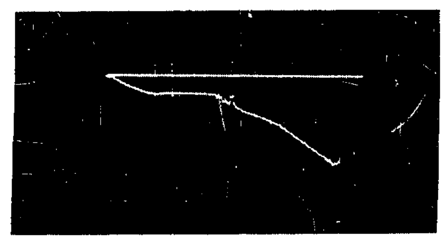


(c)



(d)

Figure 5 (a). - *Charge configuration used to compare the pressure probe system with the ionization probe system.* A barrier introduces a detonation delay time.

(b). - Record obtained by dual-beam oscilloscope display; the time base is common to both traces, the upper being the pressure record and the lower the ionization record. The time increases from left to right (5 μsec./cm) and propagation is downward. Slopes are not identical because of different lineal wire resistances.

(c). - Oscillogram obtained from the pressure probe on the same time base as (b) but with an increased vertical sensitivity (3.1 cm/div.).

(d). - Same as (c) but a record obtained from the ionization probe with a vertical sensitivity (3.9 cm/div.).

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